IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants : Hideyasu MATSUMURA et al.

Appl. No. : 10/540,866

I.A. Filed : March 22, 2004

Confirmation No. 4013
Group Art Unit : 1796

Examiner : Winkler, Melissa A

For : EXPANDABLE RESIN BEADS OF STYRENE-MODIFIED, STRAIGHT CHAIN, AND LOW-DENSITY POLYETHYLENE, PROCESS FOR THE PRODUCTION THEREOF, PRE-EXPANDED BEADS AND EXPANDED MOLDED ARTICLE

REPLY BRIEF UNDER 37 C.F.R. § 41.41(a)(1)

Commissioner for Patents
U.S. Patent and Trademark Office
Customer Service Window, Mail Stop Appeal Brief - Patents
Randolph Building
401 Dulany Street
Alexandria, VA 22314

Sir:

This Reply Brief is in response to the Examiner's Answer mailed December 12, 2008, the period for reply extending until February 12, 2009.

In the Examiner's Answer all grounds of rejection set forth in the final rejection are maintained.

Appellants note that the Examiner's Answer does not sufficiently address several of Appellants' arguments as to why the rejections are without merit, and misrepresents some of the facts. These deficiencies have prompted the present Reply Brief.

Appellants also note that this Reply Brief is being filed under 37 C.F.R. § 41.41(a)(1) and is directed to the arguments presented in the Examiner's Answer, and therefore must be entered unless the final rejection is withdrawn in response to the instant Reply Brief.

In order to avoid repetition, the following response to the Examiner's arguments in the Examiner's Answer will be limited to issues which are important enough to warrant a further comment in Appellants' opinion. Accordingly, Appellants' silence with respect to any allegations set forth in the Examiner's Answer which are not specifically addressed below should by no means be construed as Appellants' admission that these allegations are of any merit.

REPLY

1. As an initial matter, Appellants note that in the last paragraph of page 2 of the Examiner's Answer it essentially is alleged that the summary of the claimed subject matter section contained in the Appeal Brief filed September 18, 2008 is deficient because it does not mention the dependent claims which have been argued separately, as allegedly required by 37 CFR 41.37(c)(1)(v).

Appellants respectfully disagree with the Examiner in this regard. Specifically, it is pointed out that 37 CFR 41.37(c)(1)(v) refers to dependent claims which have been argued separately <u>only in the context of claims which contain means plus function and step plus function language and 35 U.S.C. 112, sixth paragraph.</u> Since the present separately argued dependent claims do not contain means plus function or step plus function language they are not required by 37 CFR 41.37(c)(1)(v) to be mentioned in the summary of the claimed subject matter section of the Appeal Brief.

2. Before addressing specific allegations in the Examiner's Answer it is deemed expedient to summarize some of the important features of the claimed subject matter and the unexpected advantageous results associated therewith.

Present independent method claims 1 and 3 have in common, *inter alia*, that they recite polymerization temperatures of (T-15) to (T-8) °C or (T+1) to (T+5) °C (where T °C is the melting point of the low-density polyethylene-based resin beads employed), 0.1 to 0.9 parts by weight of a polymerization initiator relative to 100 parts by weight of styrene-based monomer employed, and a gel component comprising less than 2 wt% of a graft polymer of the expandable beads obtained by these processes. Independent claim 5 which is drawn to expandable resin beads as such also recites a gel component comprising less than 2 wt% of a graft polymer. As set forth in the present specification, a low gel fraction of the resin beads is desirable because a high gel fraction of the resin beads renders the recycling of an expanded molded product made from these beads difficult. In this regard, page 1, line 15 to page 5, line 5 and in particular, page 4, line 13 to page 5, line 5 of the present specification may, for example, be referred to.

The present inventors have surprisingly found that the polymerization of a styrene-based monomer in the presence of linear low density polyethylene-based resin beads results in expandable polystyrene-modified beads which (a) have a gel component (gel fraction) comprising polystyrene graft copolymer of less than 2 wt% (which is advantageous, e.g., for later recycling) and (b) provide an expanded molded article which exhibits sufficient toughness of polyethylene and sufficient stiffness of polystyrene (only) if a method of the type described in JP 1-284536 to Takamasa et al. (hereafter "TAKAMASA") is carried out under certain reaction conditions, i.e.,

(i) by conducting the polymerization of the styrene-based monomer within the temperature range of from (T-15) to (T-8) °C or from (T+1) to (T+5)

°C (where T °C is the melting point of the low-density polyethylene-based resin beads), and

(ii) by using a lower amount of polymerization initiator than that taught by TAKAMASA, i.e., 0.1 to 0.9 parts by weight of polymerization initiator relative to 100 parts by weight of styrene-based monomer instead of 1.0 to 3.0 parts by weight of polymerization initiator relative to 100 parts by weight of vinyl-aromatic monomer as taught by TAKAMASA.

In this regard see, e.g., page 5, line 8 to page 6, line 2 of the present specification.

The above-noted advantageous and unexpected effects which are associated with the methods recited in present claims 1 and 3 are illustrated by the results reported in the Examples and Comparative Examples of the present specification. In this regard, reference is made to the discussion at pages 14 and 15 and page 16, first paragraph of the Appeal Brief.

The Examiner essentially maintains the position that TAKAMASA, while not disclosing the above elements (i) and (ii), allegedly renders these elements - and inherently also the amount of gel component resulting therefrom - obvious to one of ordinary skill in the art. In this regard, Appellants note that TAKAMASA does not even mention the gel component of the polymer beads described therein, let alone teaches or suggests a gel component as recited in the present independent claims.

3. Turning now to the Examiner's response to the arguments set forth in the Appeal Brief, the Examiner concedes that the polymerization temperature disclosed in TAKAMASA, (T-7) °C, is outside the range recited in present claims 1 and 3 (see, e.g.,

page 17, last sentence of next-to-last paragraph of the Examiner's Answer) but essentially alleges that there might have been a fluctuation of the melting point T°C of the polyethylene beads employed by TAKAMASA (disclosed therein to be 122°C) from batch to batch and, additionally, a variation in the polymerization temperature (disclosed to be 115°C), thereby increasing the temperature difference disclosed by TAKAMASA and thus, bringing it within the range recited in present claims 1 and 3 (see, e.g., paragraph bridging pages 17 and 18 and page 28, last paragraph of Examiner's Answer).

Appellants point out that the Examiner's allegations in this respect are unsubstantiated and unsupported by any evidence and thus amount to nothing more than mere speculation. At any rate, by the same token one could speculate that due to the alleged variations in the melting point and the polymerization temperature the actual temperature difference disclosed by TAKAMASA was (much) smaller than (T-7) °C and therefore, even further outside the ranges recited in present claims 1 and 3 than is actually disclosed by TAKAMASA.

Appellants further emphasize that in the present context it must also be kept in mind that <u>TAKAMASA neither teaches nor suggests that the difference between the melting point of the polymer beads and the polymerization temperature has any effect on the properties of the resultant beads, let alone that it can be used to adjust the amount of gel component thereof</u> and for this reason alone, is unable to render obvious the subject matter of any of the present claims.

4. In the paragraph bridging pages 18 and 19 of the Examiner's Answer it essentially is alleged that the results described and listed in Example 13 and Table 5 at

page 56 of the present specification cannot be used to support the unexpected finding that the gel fraction of the polymer beads is higher than 2 % by weight if the difference between polymer bead melting point and polymerization temperature is outside the ranges recited in claims 1 and 3 because the corresponding experiments were carried out with different catalysts. Similar allegations can be found in the first full paragraph of page 19 of the Examiner's Answer with respect to a comparison of the results obtained in Examples 1 and 4 and Comparative Examples 2-7 of the present application.

In this regard, Appellants submit that it is common knowledge in the field of polymerization that each initiator has a specific temperature range wherein it is best suited for catalyzing a polymerization, wherefore different polymerization catalysts are conventionally used (or are at least best suited) for different polymerization temperatures. In this regard, Wicher et al., U.S. Patent No. 6,608,150 (hereafter "WICHER") may, for example, be referred to.

At any rate, the results listed in Tables 1 and 2 at pages 47 and 48 of the present specification and in particular, those set forth for Examples 1 and 5 (which describe polymerizations carried out under the same temperature (and other) conditions but with different catalysts) provide evidence that the type of catalyst employed has an insignificant, if any, effect on the amount of gel fraction of the resultant resin beads.

Specifically, the polymerizations of Examples 1 and 5 were carried out at the same temperature (117 °C, T-9 °C) but with different catalysts, i.e., t-butylperoxybenzoate in the case of Example 1 and t-butyperoxy-3,5,5-trimethylhexanoate in the case of Example 5. Nevertheless, there is only a small difference in the amount of gel fraction, i.e., 0.3 % by weight in the case of Example 1 and 0.4 % by weight in the

case of Example 5, which provides evidence that the type of polymerization initiator does not affect the gel fraction to any significant extent.

5. In the passage bridging pages 18 and 19 of the Examiner's Answer the Examiner further alleges that "appellant has provided no data showing unexpected results between T-8 and T-7 (in Example 13, respectively 118°C and 119°C). Additionally, Example 13 shows the specific polymerization temperature taught by [TAKAMASA] (115°C) produces a gel content in the claimed range."

Appellants respectfully submit that these allegations are clearly without merit. For example, the results listed in Table 5 at page 56 of the present specification show that at a polymerization temperature of 118°C (<u>T-8</u> °C) the gel fraction is <u>1.8 %</u> by weight and at a polymerization temperature of 119°C (<u>T-7</u> °C) the gel fraction is <u>6.5 %</u> by weight, i.e., <u>more than 350 % of the gel fraction obtained with (T-8) °C.</u> (It is noted that in both cases the same polymerization initiator, tert-butylperoxybenzoate, was used).

Moreover, the Examiner's attention is directed to the fact that it is not an <u>absolute</u> polymerization temperature that is recited in present independent claims 1 and 3, but a <u>temperature difference</u>, i.e., the difference between the melting point of the polymer beads and the polymerization temperature. Accordingly, due to the difference in the melting points of the polymer beads employed (122°C in the case of TAKAMASA and 126°C in the case of the Examples of the present application), the fact remains that the polymerization temperature of 115°C disclosed by TAKAMASA corresponds to (T-7) °C, i.e., <u>outside</u> the ranges recited in claims 1 and 3, whereas a temperature of 115°C employed in the Examples of the present application corresponds to (T-11) °C, i.e., <u>inside</u> the ranges recited in claims 1 and 3. Accordingly, there clearly is no point in concluding

that the polymerization temperature disclosed by TAKAMASA necessarily affords the same results as those which are disclosed in the present specification for the same (absolute) polymerization temperature.

6. With respect to the comments at page 20, first paragraph of the Examiner's Answer, Appellants note that regardless of whether or not a comparison of the results in Comparative Example 2 with those in Comparative Examples 8 and 9 of the present application shows that the amount of initiator is critical for obtaining beads with satisfactory strength properties, the fact remains that according to present independent claims 1 and 3 the amount of initiator per 100 parts by weight of styrene-based monomer is from 0.1 to <u>0.9</u> parts by weight, whereas TAKAMASA specifically teaches and even <u>recites in claim 1 thereof</u> a corresponding amount of <u>at least 1.0</u> parts by weight, i.e., an amount which is <u>at least 11 % higher</u> than that recited in present claims 1 and 3.

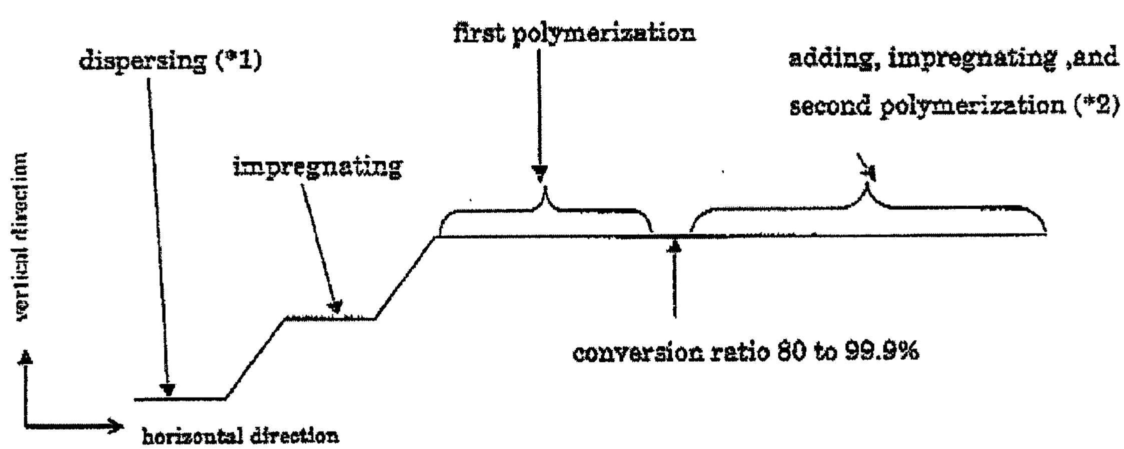
Moreover, TAKAMASA also teaches that a <u>preferred</u> amount of initiator is from 1.1-2.0 parts by weight, i.e., <u>at least 22 % higher</u> than that recited in present claims 1 and 3. TAKAMASA limits the amount of polymerization initiator to at least 1.0 (and preferably at least 1.1) parts by weight of polymerization initiator relative to 100 parts by weight of vinylaromatic monomer because this minimum concentration is necessary to obtain a favorable distribution, molecular weight distribution and size of the polymerized monomer particles inside the polyethylene beads (see, e.g., page 2, fifth paragraph and page 3, next-to-last paragraph of the machine translation of TAKAMASA).

Appellants point out that the Examiner has not provided any explanation whatsoever as to why <u>despite the clear teaching to the contrary in TAKAMASA</u> one of

ordinary skill in the art would have had an <u>apparent reason</u> to reduce the minimum amount of initiator taught by TAKAMASA by at least 10 %.

7. Appellants note that the method recited in present independent claim 3 differs from the method of recited in present independent claim 1, *inter alia*, in that in the former method the styrene-based monomer is not used in one portion (as in the method of TAKAMASA) but in two portions. In view thereof it is deemed expedient to illustrate the differences between the method of present independent claim 3 and the methods disclosed in the two documents which are relied on in all of the rejections of claims 3, 8, 13 and 16, i.e., TAKAMASA and WICHER.

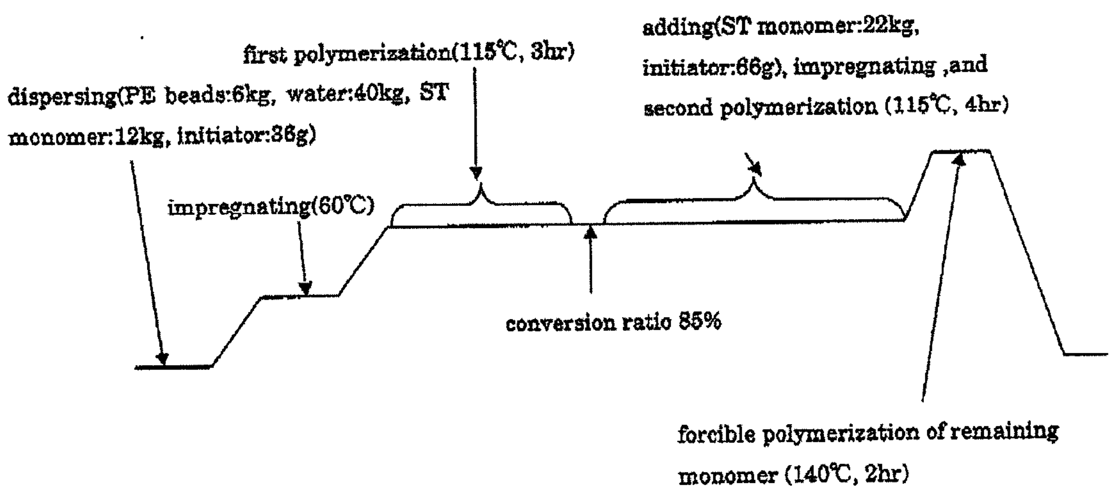
Specifically, in a temperature-time variation graph, the method recited in present claim 3 can be illustrated as follows (for the case where the temperatures in the first polymerization and the second polymerization are the same):



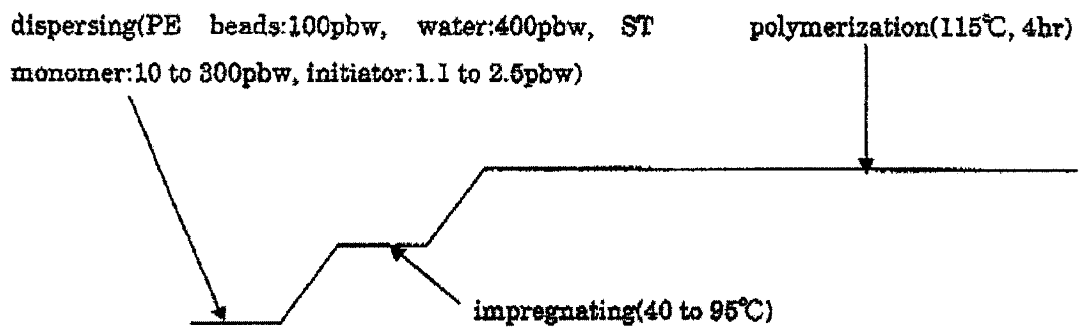
(*1) PE-based beads:100pbw
1st ST-based monomer:30 to 300pbw
initiator:0.1 to 0.9pbw/1st ST-based monomer 100pbw

(*2) 2nd ST monomer:
initiator:0.1 to 0.9pbw/1st ST based monomer 100pbw

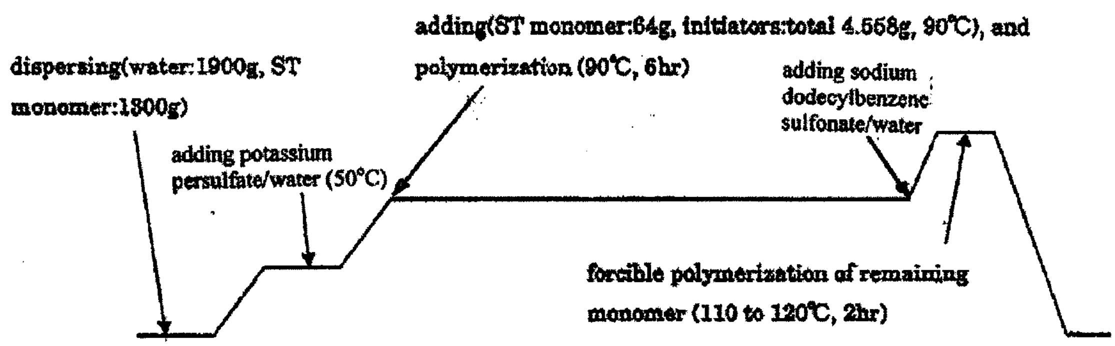
For example, the specific method described in Example 2 of the present specification (pages 31-32) can be represented as follows:



In comparison, the temperature-time variation described in the Examples of TAKAMASA can be represented as follows:



Further, the temperature-time variation described in the Example 4 of WICHER specifically relied on by the Examiner can be represented as follows:



A comparison of the above graphs makes it evident that (a) the methods of TAKAMASA and WICHER differ fundamentally in several respects, and (b) even a combination of the methods of TAKAMASA and WICHER would not result in the method recited in present claim 3 (even if the different differences between the melting points of the polymer beads and the polymerization temperature between TAKAMASA and present claim 3 were disregarded).

Specifically, the methods of TAKAMASA and WICHER differ, *inter alia*, in that the starting materials of the method of TAKAMASA are (preformed polyethylene) resin

beads and polymerizable vinylaromatic monomer (e.g., styrene), whereas the only starting material of the method of WICHER is a polymerizable monomer such as styrene and the like. Due to this difference, the method of WICHER differs from the method of TAKAMASA also in that it does not include an impregnation step (there is no solid that can be impregnated with the polymerizable monomer).

Even further, the polymerization of TAKAMASA is carried out at a single temperature and with a single catalyst, whereas the polymerization of WICHER is carried out at two different temperatures and with (at least) two different catalysts of different thermal stability.

Additionally, TAKAMASA requires the use of at least 1.0 parts by weight of polymerization initiator per hundred parts by weight of vinylaromatic monomer, whereas in all of the Examples of WICHER the total amount of the mixture of polymerization initiators employed is significantly lower, i.e., less than 0.5 parts by weight per hundred part by weight of styrene.

Still further, the product of the method of TAKAMASA are polyethylene beads which have polystyrene incorporated therein, whereas the product of the method of WICHER are (homogeneous) beads made of only one kind of polymer such as, e.g., polystyrene beads.

Appellants submit that in view of at least all of the fundamental differences between the methods of TAKAMASA and WICHER which are set forth above there is no apparent reason for one of ordinary skill in the art to combine the teachings of these documents, let alone in the manner contemplated by the Examiner.

It further is to be pointed out that neither the method of TAKAMASA nor the method of WICHER includes the polymerization of a polymerizable monomer <u>followed by the addition of additional monomer</u> and a second polymerization and for this reason alone, even a combination of the methods of TAKAMASA and WICHER in the way contemplated by the Examiner would not result in the method recited in present claim 3.

Additionally, as pointed out above in section 6., TAKAMASA fails to teach or suggest using <u>0.1 to 0.9</u> parts by weight of polymerization initiator relative to 100 parts by weight of styrene-based monomer as recited in present claim 3 but limits the amount of polymerization initiator to <u>1.0 to 3.0</u> (and preferably <u>1.1 to 2.0</u>) parts by weight of polymerization initiator relative to 100 parts by weight of vinylaromatic monomer.

For at least all of the foregoing reasons, there is no apparent reason for one of ordinary skill in the art to modify TAKAMASA by the teaching of WICHER, and even if there were, a combination of TAKAMASA and WICHER would not result in the method of present claim 3.

8. Appellants note that, e.g., in the first full paragraph of page 26 of the Examiner's Answer the Examiner maintains the position that Example 4 of WICHER discloses a two-step polymerization process wherein a first polymerization of styrene monomer at a first, lower temperature is followed by the addition of styrene monomer and a second polymerization of the styrene monomer at a second, higher temperature.

Appellants again respectfully (and strongly) disagree with the Examiner in this regard. In particular, it is apparent that the polymerizations of styrene monomer at different temperatures which are described in Example 4 of WICHER are conducted

without any addition of additional styrene monomer between these polymerizations. In particular, it is entirely clear from Example 4 of WICHER that the polymerization described therein commenced only after the entire employed styrene monomer was present, because the second portion of the styrene monomer was added together with the polymerization initiators.

Specifically, according to Example 4 of WICHER 1300 g of styrene monomer was suspended in 1900 g of deionized water containing 10.25 g of tricalcium phosphate (as suspension stabilizer, see col. 7, lines 11-13 of WICHER). The resultant suspension was heated to 50°C and 0.021 g of potassium persulfate in 50 g of deionized water was added, whereafter the resultant blend was heated to 90°C. At this temperature, i.e., before the polymerization of the styrene monomer inside the styrene droplets, three organic polymerization initiators were added to the suspension in the form of a solution in styrene monomer (64 g, less than 5 % of the suspended styrene monomer already present). Apparently, only after the polymerization initiators had been added, the actual polymerization of the styrene monomer (at two different temperatures, i.e., six hours at 90°C and two hours at 110° to 112°C) could take place.

In other words, once the actual polymerization of the styrene monomer was initiated by adding the polymerization initiators to the suspension of styrene monomer in water no additional styrene monomer was added at any point during the polymerization of WICHER, contrary to what is alleged by the Examiner. The only chemical which was added during the polymerization (before raising the temperature to the higher temperature) was suspending agent for the already formed polystyrene beads, i.e., sodium dodecylbenzenesulfonate in water.

At any rate, given the fact that according to WICHER the polymerization at the second (higher) temperature is to complete the decomposition of the organic peroxide initiators in the suspension and to provide complete polymerization of the styrene monomer (see, e.g., col. 3, line 66 to col. 4, line 3 of WICHER), it would clearly not make any sense to add fresh unpolymerized styrene monomer before this polymerization completion step.

In this regard, Appellants note that the polymerization completion step of WICHER (i.e., the polymerization at higher temperature) may be considered to correspond to the (optional) final step of the method of claim 3 which is described, e.g., in Example 2 of the present application (graphically represented above). In particular, as set forth at the end of the first paragraph of page 32 of the present specification, by raising the temperature to 140°C (from 115°C employed in both the first and the second polymerization) and maintaining the beads at this temperature for 2 hours, the remaining monomer was forcibly polymerized to reduce its amount.

9. In the passage bridging pages 24 and 25 of the Examiner's Answer it is alleged that WICHER teaches "it would have been obvious to carry out the polymerization of styrene monomers taught by [TAKAMASA] in two steps. The motivation to do so would have been that the two-step process provides advantages such as isolating the polymer beads and segregating them by size prior to a separate impregnation operation. This, in turn, allows for more precise control of bead size which is critical in some polymer molding operations". In this regard, the Examiner's Answer refers to col. 1, lines 57-60 of WICHER. Appellants note that WICHER states in col. 1, lines 48-65:

> EPS [Expandable PolyStyrene], as prepared in the suspension process, is in the form of essentially spherical beads with typical diameters of approximately 0.2 mm to 2.0 mm. In order to render the beads "expandable", it is necessary to impregnate the polymer with a blowing agent, most often low molecular weight alkane hydrocarbons like butane, 2-methylbutane, pentane and cyclohexane. EPS can be prepared in a one-step process or a two-step process. The former allows blowing agent impregnation during the polymerization and has the obvious advantage of reduced polymer handling operations. The two-step process isolates the polymer beads and segregates by size prior to a separate impregnation operation. The advantage in this case is that of precise control of bead size, a more critical parameter in some polymer molding operations. Peroxide initiator concentrations used to expedite conversion in the present invention may readily be adjusted by one of ordinary skill in the art to accommodate either process.

The Examiner appears to (incorrectly) suggest that the above passage of WICHER teaches that only with the two-step process is it possible to isolate the polymer beads and segregate them by size prior to a separate impregnation operation.

Appellants submit that one of ordinary skill in the art will understand that the isolation and segregation by size prior to an impregnation operation with blowing agent is, of course, also possible in the one-step process, provided one is willing to forfeit the advantage of reduced polymer handling operations and impregnates the polymer beads with blowing agent not already during but only after the polymerization, like in the two-step process.

At any rate, the situation is different with the process of TAKAMASA in that in this case not only unpolymerized vinylaromatic monomer but also polyethylene <u>beads</u> are a starting material and thus, a size separation of the beads which are to be impregnated with the monomer can be carried out already before any impregnation with, and polymerization of the vinylaromatic monomer inside the beads is conducted.

Accordingly, even if one were to assume, *arguendo*, that WICHER teaches a two-step polymerization with the employment of additional monomer between the two

polymerization steps and teaches that the two-step process has advantages compared to the one-step process with respect to size separation of the beads, it is not seen that WICHER provides any apparent reason for one of ordinary skill in the art to use this (assumed) technique in the method of TAKAMASA.

5. In the paragraph bridging pages 30 and 31 of the Examiner's Answer it is alleged that both ends of the polymerization temperature range disclosed in co-pending Application No. 10/541,391 are "remarkably close" to the upper and lower ends of the temperature ranges recited in the present claims, and that the claimed gel content in the claims of Application No. 10/541,391 is "very close" to the gel content recited in the present independent claims. It further is alleged that "the claimed methods and products thus appear to be modifications of each other derived from routine experimentation and optimization" and that "[p]rocessing conditions, such as temperature, are routinely adjusted for reasons such as enhancing speed or reducing cost".

Appellants respectfully submit that while it may be that in cases where no or only a very broad value range for a particular parameter is indicated in a document one of ordinary skill has a reason to conduct experiments in order to find a workable or optimum value range. However, this is not the case here. The value range disclosed in Application No. 10/541,391 spans only a few degrees centigrade and, moreover, is recited in the claims thereof, which is an indication for one of ordinary skill in the art that this range was considered important by the inventors of the co-pending application. Accordingly, there is no reason for one of ordinary skill in the art to carry out routine experimentation and optimization in the present instance.

CONCLUSION

The request to reverse the rejection of claims 1-16 and to return the application to the Examining Group for prompt allowance is respectfully maintained for at least all of the foregoing reasons.

Although no fee is believed to be required for entry of this Reply Brief, the Patent and Trademark Office is hereby authorized to charge any fee that is deemed to be necessary to Deposit Account No. 19-0089.

Respectfully submitted,
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